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19. ABSTRACT (Continue on reverse if necessary and identify by block number)

Viscoelastic materials with fading memory, e.g. polymers, suspensions, emulsions, exhibit behavior that is intermediate between the nonlinear hyperbolic response of purely elastic materials and the strongly diffusive, parabolic response of viscous fluids. The following problems that are an outgrowth of earlier research supported by ARO were investigated during the reporting period:

(1) Provide numerical and analytic explanation of several striking phenomena observed in experiments in shear flows of highly elastic and very viscous non-Newtonian fluids in pressure-driven and piston-driven flows and in Couette flow. Such phenomena can lead to material instabilities deemed capable of severely disrupting advanced materials engineering and process design problems of polymer melts that include spinning of synthetic fibers and injection molding.

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18. SUBJECT TERMS - cont'd.

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19. ABSTRACT - cont'd.

2. Establish the nonlinear stability of possible discontinuous, steady non-Newtonian shear flows in order to justify various approximations made in the analysis of the nonlinear partial differential equations (PDE) governing the unsteady motions studied under (1).

3. Prove the existence of weak solutions to a system of a first-order quasilinear hyperbolic system of Volterra integrodifferential equations modelling the unsteady motion of a viscoelastic bar with fading memory for bounded initial data of arbitrary size.

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Interdisciplinary Research on Viscoelasticity & Rheology

Final Report

John A. Nohel
December 14, 1990

for the
U.S. Army Research Office
DAAL03-87-K-0036

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A. STATEMENT OF THE PROBLEMS STUDIED: Viscoelastic materials with fading memory, e.g. polymers, suspensions, emulsions, exhibit behavior that is intermediate between the nonlinear hyperbolic response of purely elastic materials and the strongly diffusive, parabolic response of viscous fluids. Their behavior is influenced by a subtle dissipative mechanism induced by effects of the fading memory; a fundamental understanding of the equations governing their unsteady motions under physically relevant constitutive assumptions is crucial for modelling, analysis, design of algorithms and computation of specific problems.

The following problems that are an outgrowth of earlier research supported by ARO were investigated during the reporting period:

(1.) Provide numerical and analytic explanation of several striking phenomena observed in experiments in shear flows of highly elastic and very viscous non-Newtonian fluids in pressure-driven and piston-driven flows and in Couette flow. Such phenomena can lead to material instabilities deemed capable of severely disrupting advanced materials engineering and process design problems of polymer melts that include spinning of synthetic fibers and injection molding (see, e.g., M. M. Denn *Ann. Rev. Fluid Mech.* 22 (1990), 13-34.)

(2.) Establish the nonlinear stability of possibly discontinuous, steady non-Newtonian shear flows in order to justify various approximations made in the analysis of the nonlinear partial differential equations (PDE) governing the unsteady motions studied under (1.)

(3.) Prove the existence of weak solutions to a system of a first-order quasilinear hyperbolic system of Volterra integrodifferential equations modelling the unsteady motion of a viscoelastic bar with fading memory for bounded initial data of arbitrary size.

Our approach to resolving these problems was guided by the following objectives:

- i. To understand qualitative properties of the mathematical model: the global well-posedness of solutions to the governing PDE, their regularity and asymptotic behavior for large time, approach to steady states, etc.
- ii. To understand the physical consequences of the model: Do any or all of the possible solutions make physical sense? Do solutions that have mathematically interesting character correspond to observed phenomena? Do they predict behavior that should be studied in the laboratory? What solutions to the problem are relevant to processing and design?
- iii. To understand the physical model: How do the observed solutions correspond to the molecular or continuum model on which they are based? Can the character of these solutions serve to validate the physical model or suggest improvements in it?
- iv. To design numerical methods that account for the mathematics and reflect the physics.
- v. To study the broad mathematical implications of i. - iv. for these and related classes of problems. We note that concepts such as weak solutions, shocks, shear bands, phase changes, etc arise in a variety of problems of physical and technological importance.

Considerable progress and understanding of the problems has been gained using this approach. Initial numerical studies suggested scaling that led to new analytical results, approximation techniques and improvements in numerical methods that in turn provided a deeper understanding of the underlying physics and yielded a clear explanation of observed phenomena.

B. SUMMARY OF THE MOST IMPORTANT RESULTS:

This outline relies on the fact that more details are available in semi-annual reports submitted during the award period, and in the manuscripts already furnished and those being sent to ARO under separate cover; numbers in brackets refer to items listed in part C. below.

The phenomena to be understood and analysed under part (1.) were observed in three recent experiments: (a) "Spurt", a sudden large increase in volumetric flow rate observed in quasi-static

loading of pressure-driven shear flows of monodisperse polyisoprenes through a capillary (equivalently a slit-die, as shown in the Ph. D. thesis of M. Yao, see D. below) at a critical stress that is independent of molecular weight, was discovered by Vinogradov et al. (*J. Polymer Sci., A-2* **10** (1972), 1061-1084); the experimental results were confirmed by Lyngaae-Jorgensen & Marcher (*Chem. Eng. Comm.* **32** (1985), 117-151). (b) Nearly periodic disturbances were observed by Lim and Schowalter (*J. Rheology* **33** (1989)) in piston-driven channel flow of a highly elastic and viscous non-Newtonian fluid at a fixed volumetric flow rate and they were attributed to "stick-slip". (c) In the third experiment, R. Larson and F. Morrison (preprint (1989)), measure the stress response of highly elastic, viscous non-Newtonian fluids in "step-strain".

Until recently, the spurt phenomenon had been overlooked or dismissed by rheologists because no plausible mechanism was known to explain it in the context of steady flows. Spurt was lumped together with instabilities such as "slip," "apparent slip," and "melt fracture" which are poorly understood (see article by M. Denn cited above). The mechanism that induces spurt was not understood because popular numerical methods, based on steady viscoelastic fluid flows falter in "high Weissenberg number" regimes and thus cannot model the spurt phenomenon. Our research indicates that spurt and related phenomena can be modeled and explained by studying the full dynamics of the unsteady equations of motion and constitutive equations.

The systems of time-dependent quasilinear PDE's used to model all three experiments under incompressible and isothermal conditions are derived in [10], [12], [15], [23], [21] in a fully three-dimensional setting. The total shear stress is decomposed into a contribution due to the polymer, evolving in accordance with a Johnson - Segalman - Oldroyd (JSO) differential constitutive law (with single and also multiple relaxation times) where the differential operator is objective, a small Newtonian contribution and a pressure term. For a Newtonian fluid the governing system reduces to the Navier-Stokes equations. A major emphasis during the grant period was placed on understanding spurt and related phenomena discovered in extensive numerical simulation; analysis of piston-driven and step-strain flows was initiated during the past year.

Major results obtained for the problems in (1.) are:

a. Pressure-driven unsteady flow through a narrow slit-die is modelled by an initial boundary value problem for shear flow of a non-Newtonian fluid between parallel plates, symmetric about the centerline and with initial data consistent with the no-slip boundary conditions at the wall. The unknown flow variables are the velocity distribution, the polymer shear stress and a quantity proportional to the first normal stress difference. For Vinogradov-type materials modelled by (JSO) with a single relaxation time the dynamic response involves multiple time scales; it is shown in [12] and [15] that there are two essential dimensionless flow parameters: α , the ratio Reynolds to Deborah or Weissenberg numbers, and ϵ , the ratio of Newtonian to shear viscosities. In [15], we used asymptotic analysis for a model with two widely separated relaxation times and no Newtonian viscosity to show that the dynamics of the two models are similar.

b. If $\epsilon = 0$, the governing system can change type (from strictly hyperbolic to elliptic [2]). For smooth initial data in the strictly hyperbolic region, energy methods described in [2] can be used to show that smooth solutions exist globally in time if the data are sufficiently small. By contrast, the method of characteristics developed for viscoelastic problems in [1], [2] yields that initially smooth solutions exhibit finite-time blow-up (of derivatives) if the initial data are sufficiently large. If $\epsilon > 0$, the governing system is evolutionary but cannot be classified as to type. General theory developed in [16] for semilinear reaction-diffusion type parabolic systems to which the governing system can be transformed yields global-time existence and uniqueness of classical solutions for smooth initial data of arbitrary size, and also existence and uniqueness of global, almost classical, strong solutions with discontinuities in the initial velocity gradient and stress components. Although globally well-posed, it is difficult to prove that solutions to the governing system achieve steady states as $t \rightarrow \infty$.

However, it is a simple matter to find the steady states and show that the total steady shear stress is a nonmonotone function of strain rate (for relevant ϵ) leading to multiple steady states that suggest the possibility of existence of steady solutions having finite jumps in strain rate and existence of steady velocity profiles with "kinks" (jump discontinuities in the steady velocity gradient) [10], [12], [15].

c. Three distinct numerical algorithms for the governing time-dependent system were developed in [6], [10], [12]: The first is based on a stable, finite-element, solid-mechanics method originally developed by Malkus [6], [11], [20] in which the system is regarded as governing extensional motion of an elastic-plastic bar. The second uses the equivalent reaction-diffusion system and is based on solving a forced, nonlinear heat equation coupled with nonlinear ODE's. The third uses the fact that the governing system can be transformed to conservation form [10], [12], and the numerical solution is obtained by using techniques of hyperbolic conservation laws with a regularizing Newtonian viscosity and is based on the solution of the Riemann problem when $\epsilon = 0$; steady numerical solutions are achieved by wave propagation and wave interactions. The third of these is the only method capable of yielding satisfactory numerical solutions even in the absence of Newtonian viscosity [12]. Calculations with these methods produced similar qualitative and quantitative results.

d. Detailed numerical simulation of the Vinogradov quasistatic loading experiment at a very small Reynolds number using the solid mechanics algorithm and a code developed by Malkus is explained in [12]. The simulation produced excellent qualitative and quantitative agreement with experimental results for all eight material samples that differ from one another only in molecular weight. For samples relevant to polymer processing, the Weissenberg (Deborah) number is very large. The dimensionless parameters α, ϵ used in the model are both very small with α seven to ten orders of magnitude smaller than ϵ . The numerical solution exhibits discontinuities when steady state is achieved and indicates that spurt occurs at a critical value of the total stress that is independent of molecular weight (near the shear stress maximum of the steady equations) as the driving pressure gradient is raised from values slightly below to slightly above critical; the dramatic increase in the volumetric flow rate corresponds to a jump in the steady strain rate at the local maximum which leads to a steady velocity profile with kink at a narrow layer near the wall. However, much more is revealed: There is also a local minimum in the steady shear stress, and further numerical simulation in [12] suggests that unloading leads to a hysteresis loop whose magnitude is governed by the difference between the stress at the local maximum and the local minimum. The simulation also revealed "latency" (period preceding spurt), "shape memory" under cyclic loading and unloading, as well as behavior under flow reversal. This and other numerical testing showed that *the common key feature of differential constitutive models that capture these phenomena is a non-monotonic relation between the steady shear stress and strain rate*. Therefore, at least for these flows, our approach to finding numerical solutions of the unsteady problems successfully overcomes the "high Weissenberg number" problem that often plagues numerical simulation of corresponding steady viscoelastic flows. To provide further credence to the mathematical model, new experimental procedures are suggested in [12] to verify latency, shape memory, and width of the hysteresis loop.

e. To explain results of numerical simulation analytically, we use the approximation that α is nearly zero for highly elastic, very viscous materials such as the polyisoprenes in the experiments of Vinogradov. For every value of the driving pressure gradient, the governing system of PDE's is then reduced to a new, one-parameter family of systems of two autonomous, first-order quadratic ODE's, one for each point in the channel. There is a one to one correspondence between steady states of the PDE's and critical points of the ODE's. The complete dynamics of this new quadratic system has a rich structure that is determined completely in [12] - [15] by nontrivial application of

phase plane and asymptotic techniques. The salient features of the quadratic system are:

- i. Whenever the pressure gradient is such that the total steady shear stress falls between the local maximum and minimum, two of the the three resulting critical points are locally asymptotically stable, while the third is a saddle; saddle-node bifurcations occur at the maximum and minimum. For values of the total stress above the maximum or below the minimum, the single critical point is locally an attractor.
- ii. There are no periodic or homoclinic (separatrix) cycles.
- iii. While the quadratic system does not define a gradient flow, a Lyapunov-like energy relation enables one to determine invariant regions.
- iv. As a consequence, we determined the location of the stable and unstable manifolds of the saddle and the basin of attraction of each asymptotically stable critical point. *Globally, every solution of each quadratic system tends to a unique critical point as $t \rightarrow \infty$; correspondingly, when $\alpha = 0$, every solution of the governing system tends to a unique steady state.*

f. The analytical results summarized in e. explain the occurrence of spurt and hysteresis under cyclic loading and unloading [15]. When combined with additional asymptotic analysis [15], they also yield two rather dramatic effects that were observed in the numerical solutions: latency and shape memory. Roughly speaking, latency is the tendency of the polymer system to have the appearance of steady, classical Poiseuille flow during a (possibly) long period during which velocities and shear stresses do not change, while normal stresses change until a point of catastrophic transition is reached and spurt occurs. The analysis also identified the parametric dependence of latency and made precise predictions of what experimental evidence would be a signature of this phenomenon. Shape memory was found to relate to the extent of the basin of attraction of a "spurting" critical point (a globally attracting spiral point that lies above the local shear stress maximum).

The remainder of part (1) summarizes work in progress on piston-driven and step-strain flows related to the Lim-Schowalter and Larson-Morrison experiments and remarks numerical techniques for more complex flows.

g. To model the seemingly periodic disturbances observed in piston-driven shear flow of highly elastic, viscous non-Newtonian fluids at fixed volumetric flow rate, we use recent work by Deiber and Schowalter (private communication) showing that the data of Lim-Schowalter are consistent with the assumption of non-monotonicity of the total steady shear stress vs. strain rate curve. We model the flow by the initial-boundary value problem described in a. above with the additional constraint that the pressure gradient is to be determined to keep the volumetric flow rate constant. The resulting nonlinear feedback control problem is globally well-posed in time [23]. Taking $\alpha = 0$ (for the reasons described in d. above) and finding the feedback control provides a reasonable approximating system in the form of a new *quadratic system of functional differential equations* (QFDE) that is also globally well posed. Our results to date in [23] are:

- i. The linearization of (QFDE) has a complicated spectrum; there is a continuous component and the point spectrum is difficult to determine even numerically.
- ii. Numerical simulation of the Lim-Schowalter experiment using an algorithm for solving (QFDE) developed by Malkus is encouraging. The numerical solutions appear to be very nearly periodic over a time interval that corresponds to the experiment, with an apparent period consistent with experimental observations.
- iii. Numerical simulation also suggests how (QFDE) can be approximated further by a globally well-posed system of four first-order quadratic, autonomous ODE's; its dynamics is complicated! The three critical points and their local nature can only be found numerically (done by two different methods). For appropriate data the numerical solution of the system of ODE's decays very slowly as $t \rightarrow \infty$ in a nearly periodic fashion (consistent with the experiment and

the numerical simulation of (QFDE)) to a unique critical point. Since the 4-dimensional flow is not gradient-like, it is extremely difficult to rule out periodic and homoclinic orbits and to determine the global dynamics by known analytic methods.

Results in [23] obtained to date raise the possibility that the appearance of stick-slip flow (asserted by Lim-Showalter) can be emulated by spurt and may derive from the same physical mechanism. This possibility was raised in the review article by M. M. Denn cited above.

h. The step-strain experiment for a highly elastic, viscous non-Newtonian fluid is modelled as planar unidirectional Couette flow between parallel plates in which the lower wall is fixed and the upper wall is moved suddenly at $t = 0$. Using the JSO constitutive law leads to a nonlinear initial-boundary value problem similar to that described for pressure-driven flow in a. above, with no pressure term, zero initial data, and with an inhomogeneous boundary condition that measures the strain at the upper plate [21]; the problem is globally well-posed in time. Numerical solution by Malkus and Kolkka of the dynamic governing system using Vinogradov's material data, shows that at a sufficiently high average strain, a spurt-like inhomogeneous flow develops while the stresses are relaxing and the initial flow has essentially ceased. The stress response in the numerical simulations agrees very well at moderate strains with the Morrison-Larson experiments that used a different material; there is disagreement at higher strains. There are plans to redo the experiment using Vinogradov type materials.

To gain deeper insight, a formal, composite singular perturbation expansion of the solution to the governing system for step-strain in powers of the parameter α is found in [21] using initial layer corrections. When $\alpha = 0$, the reduced problem is another nonautonomous, quadratic system of functional differential equations that is globally well-posed in time; all of its solutions are bounded, and can be found numerically. Higher order terms in the expansion are found recursively and are readily computable. Spatial inhomogeneity develops at order one in α , and grows to order one in the nonlinear response, consistent with numerical simulation of the full governing system.

i. We close this section with remarks on current development of computational methods that generalize our techniques for shear flows to more complicated flows. The key feature leading to the success of the method for shear flows is that it treats the short time-scales associated with Newtonian viscosity (or short relaxation processes) and shear-wave propagation implicitly, while treating the long relaxation processes explicitly. The method has been generalized to flows with non-constant strain-rate histories in the context of the well-known fiber-drawing problem [20]. Analysis of the numerical methods shows that the ratio of short to long relaxation times or the Newtonian viscosity ratio is a key parameter in the stability and accuracy of the methods. When this is properly accounted for, the techniques described in [20] work well in shear and extensional flows and show promise for two-dimensional flows. A pressing question about two-dimensional flows being addressed is: Are steady, spurt-layers in shear flows stable to higher-dimensional disturbances? If not, are the unstable modes periodic and do they have wave numbers characteristic of known processing instabilities? We are exploring the possibility that at least a partial answer to these questions can be provided by a combination of techniques already developed for one-dimensional flows with spectral methods. This approach, of independent scientific interest, will provide a natural technical bridge to the study of more complicated, two-dimensional flows.

(2.) The results summarized in (1.)a.-f. above strongly suggest that the dynamics of the governing system of PDE's based on the JSO constitutive assumptions is very similar to that of the one-parameter system of the approximating quadratic ODE's. A significant test of this conjecture is to study the nonlinear stability of steady velocity profiles with kinks, at least when the parameter $\alpha > 0$ is small.

a. For a simpler model system of PDE's that can be arranged to have the same behavior in steady shear as JSO system, it is shown in [16], [17] via a priori estimates that independent of α ,

all solutions are bounded; the total stress tends to zero, and every solution approaches a possibly discontinuous steady state as $t \rightarrow \infty$. Moreover, discontinuous steady states taking their values on the strictly increasing parts of the non-monotone steady shear stress vs strain rate curve are nonlinearly asymptotically stable with respect to perturbations of initial data that omit sets of arbitrarily small measure surrounding each point of discontinuity in the steady strain rate. Such steady states are local minimizers of an associated, generally non-convex, stored energy functional.

b. In research in progress [22], the nonlinear stability result is being established for the governing system of PDE's based on JSO by combining geometric techniques for the equivalent semilinear, parabolic reaction-diffusion system (see (1.)b. above) with dynamical systems theory applied in suitably constructed neighborhoods of the manifold of equilibria of the one-parameter quadratic ODE's. A crucial difficulty is that a priori estimates that are essential for the variational approach in a. are no longer available. For small enough $\alpha > 0$, the statement of the new stability result is the same as in a. We expect that a delicate resolution of the saddle-node bifurcations that take place at "top" and "bottom" jumping will also yield the nonlinear stability of a "spurting" solution.

c. A formal, composite singular perturbation expansion of the solution to the governing system of PDE's for pressure-driven flow in powers of the parameter α is found in [21] using initial layer corrections. When $\alpha = 0$, the reduced problem is the one-parameter family of quadratic systems of ODE's the dynamics of which is known completely (see (1.)e.). An a priori estimate bounding the total stress by a constant (depending only on data) times α , proved in [21], implies that the formal expansion is uniformly valid at zero order; very few rigorous results for singularly perturbed systems of PDE's are available.

(3) Unsteady motions of viscoelastic solids with fading memory that are modeled well by discontinuous strains and velocity fields are observed experimentally. This fact, coupled with results concerning the formation of singularities in finite time for large smooth data ([1], [2]) suggests that one must allow for weak solutions in order to discuss global behavior of solutions for large data. Despite the importance of the subject, comparatively little is known about existence of weak solutions in the context of viscoelastic materials ([7] provides a survey of known results). In [8], [9], we discuss a model hyperbolic system of Volterra integrodifferential equations in the important special case when the two material functions appearing in the instantaneous elastic response and in the history term (in the form of a convolution integral) are equal. We use the method of vanishing viscosity and compensated compactness to obtain the global-time existence of a weak solution (in the sense of distributions) for Cauchy data that are bounded, measurable and square integrable, but are not restricted in size. If the memory kernel is identically zero, the analogous problem for one dimensional elastic motions and for motions of isentropic gases was solved in pioneering research by Di Perna ('83). The principal difficulty for the viscoelastic problem lies in establishing suitable L^∞ estimates for the regularized parabolic system that permit application of the compensated compactness method. In contrast to the situation in Di Perna's work, such estimates are difficult to achieve here by finding an invariant region because of the nonlocal nature of the memory. As for other results of this type, there is no satisfactory uniqueness theorem.

In view of continued interest in applications of Di Perna's techniques by the conservation laws community, we note that extending the results in [8],[9] to the more general viscoelastic case of two unequal nonlinear material functions presents formidable difficulties; for a model problem of a single conservation law with memory, such a result was recently obtained by Dafermos (1988) but his technique does not seem to be extendable to systems. On the one hand, assuming the existence of L^∞ estimates for solutions of an appropriately regularized system, it does not seem possible to prove convergence to a weak solution as the viscosity parameter tends to zero, because of a lack of entropies. On the other hand, it seems equally difficult to establish L^∞ estimates for solutions

of an appropriately regularized system; when the nonlinearities are different, the approach used in [8], [9] fails because the integral term can no longer be treated as a lower-order source term. The situation is essentially the same as that encountered when attempting to establish existence of weak solutions for nonisentropic gas dynamics (which alluded Di Perna and Tartar and remains an open problem).

C. LIST OF MANUSCRIPTS PUBLISHED OR SUBMITTED AND IN PREPARATION UNDER ARO SPONSORSHIP DURING THE PERIOD OF THIS AWARD INCLUDING JOURNAL REFERENCES:

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D. LISTING OF ALL PARTICIPATING SCIENTIFIC PERSONNEL SHOWING ANY ADVANCED DEGREES EARNED BY THEM WHILE EMPLOYED ON THE PROJECT:

J.A. Nohel (entire period), D.S. Malkus (through June, 1990).

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